

ABILITY FOR SEPARATE REGISTRATION BY PULSE SHAPE OF *p*-TERPHENYL SINGLE CRYSTALS ACTIVATED WITH VARIOUS CONCENTRATIONS OF 1,4-DIPHENYL-1,3-BUTADIENE

A.V. Krech, I.V. Lazarev, S.V. Makhota, E.V. Martynenko, O.A. Tarasenko

Institute for Scintillation Materials, NAS of Ukraine, Kharkiv, Ukraine

E-mail: antonkrech@gmail.com

The influence of dopant concentration on pulse shape discrimination ability of the *p*-terphenyl single crystals doped with 1,4-diphenyl-1,3-butadiene was investigated. The figure of merit (FOM) values were obtained under mixed alpha+gamma radiation for *p*-terphenyl single crystals containing different concentrations of 1,4-diphenyl-1,3-butadiene. Charge-time transformation method was used. The content of 1,4-diphenyl-1,3-butadiene in *p*-terphenyl crystals varied from 0 (pure *p*-terphenyl) to approximately 0,8 mass.%. Obtained results has shown, that with increase of introduced 1,4-diphenyl-1,3-butadiene concentration the pulse shape discrimination ability of *p*-terphenyl single crystals is getting worse in general.

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INTRODUCTION

p-Terphenyl is a widely known organic scintillation material which is successfully used in many tasks due to short decay time, high efficiency of detection of short-range radiations and also ability to selective detection of ionizing radiations by a pulse shape. The pulse shape discrimination ability of ionizing radiations of different types is especially important feature for scintillators because the effect on medium differs significantly for radiations of different types and natural isotopes are mostly sources of mixed radiation fields [1].

A scintillation pulse of an organic scintillator consists of the fast and the slow components. Pulses caused by the ionizing radiations with high specific energy losses dE/dx (alpha-particles, recoil protons, etc.) have more intensive slow component than those caused by ionizing radiations with low dE/dx (for instance, gamma-radiation, conversion electrons). Thus, it is possible to identify the type of radiation that caused the scintillation pulse by the intensity of the slow component and its relative contribution to the total scintillation pulse.

The fast component is caused by direct transition of singlet-excited states to the ground state, that is $S_1 \rightarrow S_0$ transition. The slow component is formed through the singlet channel of the intermediate process of triplet-triplet annihilation [1]:

$$T_1 + T_1 \rightarrow \begin{cases} S_1 + S_0 \rightarrow h\nu + 2S_0 \\ T_1 + S_0 \\ Q + S_0 \\ \dots \end{cases} \quad (1)$$

An efficiency of the processes of transport and recombination of triplet states determines the intensity of the slow component of a scintillation pulse and, as a consequence, the selective ability of a scintillator.

It is known, that doping of single crystal of *p*-terphenyl with 1,4-diphenyl-1,3-butadiene results in the growth of light output values, that is increasing the number of photons generated by the same type of excitation [2–4].

Increasing of light output values for doped single crystals of *p*-terphenyl (PTP) when doped with 1,4-diphenyl-1,3-butadiene (DPB) caused by the effective transfer of electronic excitation energy from the host molecules (PTP) to the molecules of dopant (DPB). For this reason, the doped crystals of PTP are more commercially demanded.

On the other hand, adding the dopant an amount that exceeds certain concentration leads to decreasing of the light output due to the concentration quenching, to loss in transparency of a scintillator and also it can significantly reduce its pulse shape discrimination ability [4–6].

The influence of DPB concentration on the light output values for scintillators of PTP doped with DPB was studied in [1, 4, 6]. At this work we investigated the influence of DPB concentration on the selective ability of PTP single crystals doped with it.

EXPERIMENTAL SAMPLES AND METHODS

The single crystal sample of pure PTP and 3 series of single crystal samples of PTP doped with DPB, containing different concentrations of DPB, were investigated. Samples that belong to the same series were obtained by sawing of the single crystal boule of PTP doped with DPB that was grown from the melt with given concentration of introduced DPB. DPB was added to the melt in the following initial concentrations: 0.05, 0.1, and 0.3 mass.%. Each series was consisted of 6 single crystal samples 2 mm thick, which were numbered from 1 to 6. Number 1 corresponds to the sample from the beginning part of the single crystal boule and number 6 – to its end part according to crystal growth direction. One can find more details about sample preparation in our previous work [6].

During the growth process of the PTP single crystal doped with DPB from a melt, the dopant is rejected by the freezing solid and is accumulated in the melt. Thus, DPB concentration in different parts of the grown single crystal boule differs, as we had showed in [6], it increases along crystal growth direction. That is, for every series of investigated samples the concentration of

contained DPB increases with sample number from 1 to 6. DPB content also increases for all samples of the series with an increase in the initial amount of DPB added to the melt (Table 1). The method of gas chromatography mass spectrometry was used to determine DPB content in the samples of PTP single crystals [6]. In this work, the pulse shape discrimination ability of the above-mentioned single crystal scintillators was determined for the case of mixed irradiation with alpha-particles and photons of gamma radiation using the charge-time transformation method.

Table 1
DPB content in the samples of PTP single crystals for different initial concentrations of DPB added in melt [6]

Sample number	PTF+0.05 mass.% DPB	PTF+0.1 mass. % DPB	PTF+0.3 mass. % DPB
1	0.0094	0.0169	0.0544
2	0.0087	0.0184	0.0685
3	0.0107	0.0178	0.0859
4	0.0161	0.0332	0.1581
5	0.0219	0.0517	0.2608
6	0.0764	0.232	0.7979

According to the above-mentioned, the scintillation pulses caused by alpha-particles have more intensive slow component than those caused by gamma-quanta. So, it is possible to identify the type of radiation that generated scintillation pulse by the relative contribution of the "tail" part of the pulse into the total pulse.

The measurements of the light transmittance T in the range from 200 to 800 nm were performed using Shimadzu-2450 spectrophotometer with the integrating sphere. The comparison channel remained blank, and the light flux inside it was calibrated to be the same as the light flux falling on a sample in the measuring channel. The inaccuracy of the calibration was limited to 0.5%. The value of T was calculated as follows:

$$T = \frac{I}{I_0} \times 100 \%, \quad (2)$$

where I_0 is the light flux in the comparison channel, I is the light flux, which has passed through a sample in the measuring channel. Actually, the T -value (2) is a relative light transmittance, where $T = 100\%$ is the luminous transmittance of air.

RESULTS

For all investigated samples, the values of transmittance (T) were measured in the range from 200 to 800 nm. Fig. 1 demonstrates examples of the obtained dependencies for some samples.

Using these experimental spectra, the dependences of transmittance at a wavelength of 390 nm on DPB content were obtained for easier perception and analysis of the information (Fig. 2). The wavelength of 390 nm was chosen because it corresponds to the luminescence wave of the scintillators under investigation.

As one can see from the results, presented in the figure, for the investigated range of concentration, a tendency for weak growth in transmittance with increasing DPB content is observed on average.

Figs. 3 and 4 show the dependences of the light output of PTP scintillators activated by DPB on the

This method allows to separate pulses caused by photons of gamma radiation and alpha-particles using differences in shapes of scintillation pulses generated by these types of radiation. This method is described in detail in [1, 7, 8]. Data were analyzed autonomously by charge comparison method (CCM) [9, 10]. Pu^{239} (alpha-particles + gamma radiation) and Cs^{137} (gamma radiation) sources were used for excitation of the samples. Cs^{137} source was in the metal container to avoid conversion electrons hit a scintillator.

DPB content for excitation by gamma radiation and alpha particles, respectively.

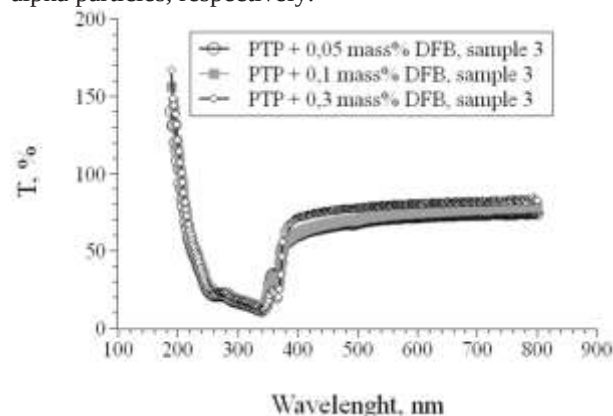


Fig. 1. Spectra of transmittance for sample 3 of series PTP+0,05 mass.% DPB, sample 3 of series PTP+0,1 mass.% DPB, sample 3 of series PTP+0,3 mass.% DPB

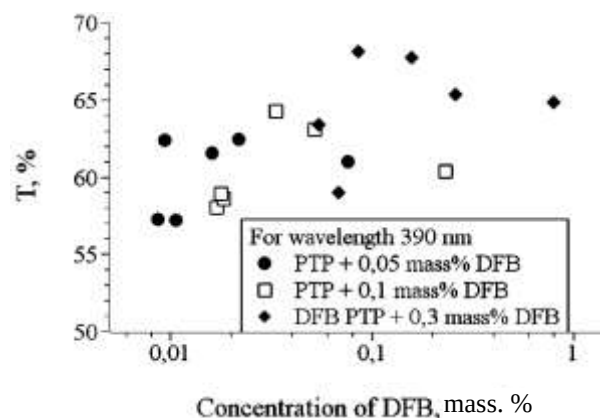


Fig. 2. Dependence of transmittance of single crystals of PTP on DPB content

These data, as mentioned earlier, were obtained in [6]. We present them in this work for the convenience

of analyzing and presenting the values of the relative light output depending on the DPB concentration in PTP for different radiations.

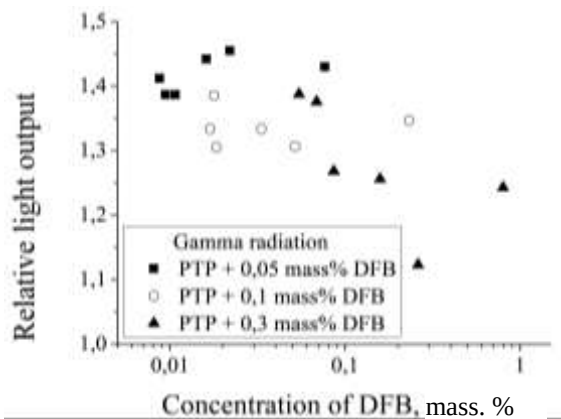


Fig. 3. Dependence of the relative light output of single crystals of PTP on DFB content under excitation with photons of gamma-radiation. The light output of pure PTP was taken as 1

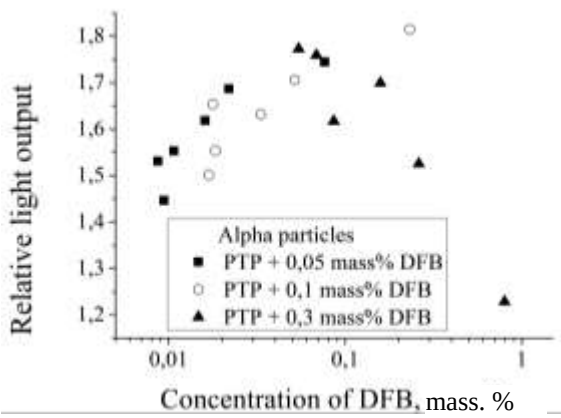


Fig. 4. Dependence of the relative light output of single crystals of PTP on DFB content under excitation alpha particles. The light output of pure PTP was taken as 1

Fig. 5 demonstrates, as an instance, spectra of relation of "tail" part area of the pulse to the total area of the pulse (tail-to-total relation) for pure PTP, sample 1 of series PTP+0.1 mass.% DPB, sample 5 of series PTP+0.3 mass.% DPB. They are typical spectra obtained with charge comparison method. The peak on the left corresponds to events caused by gamma-radiation, for pulses generated by it have lower contribution of the slow component. The peak on the right corresponds to events caused by alpha-particles.

Curves similar to those presented at Fig. 5 were obtained for all investigated samples.

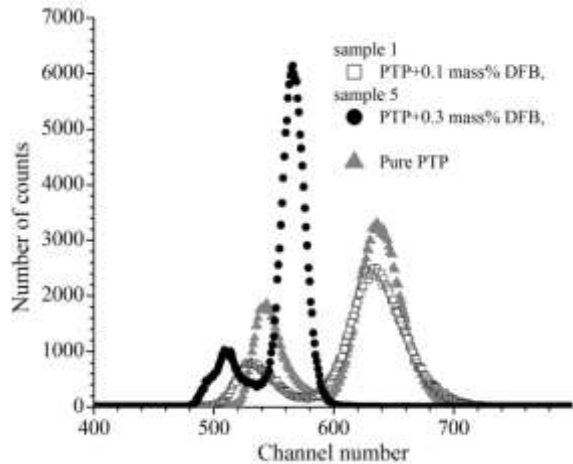


Fig. 5. Spectra obtained with charge comparison method for the sample of pure PTP, sample 1 of series PTP+0.1 mass% DPB, sample 5 of series PTP+0.3 mass.% DPB

The pulse shape discrimination ability of the scintillator was characterized using the FOM-value that is determined by:

$$FOM = \frac{\Delta A}{H_1 + H_2}, \tag{3}$$

where ΔA – is difference of maximums of alpha- and gamma peaks; H_1 and H_2 – are full widths at half maximum of alpha- and gamma peaks, respectively.

Using the obtained spectra, the FOM values were calculated for all samples by formula (3). The results of these calculations are presented in Table 2.

As one can see from the data presented in Tables 1 and 2, for the majority of samples (with the exception of 3 of them) FOM values for doped PTP single crystals were smaller than for pure one. Despite the fact that a clear dependency of FOM-values versus DPB content cannot be distinguished, the general tendency to reducing of FOM-value (that is to worsening of selective ability of PTP crystals) with DPB concentration growth is observed. For sample 6 of series PTP+0.3 mass.% DPB, which has the maximal content of DPB among all samples under investigation, it was unable to obtain FOM value because peaks of spectrum of relation of "tail" part area of the pulse to the total area of the pulse were not resolved.

Table 2

The FOM-values for the single crystal of pure PTP and the single crystals of PTP grown from melt with adding 0.05, 0.1, and 0.3 mass.% of DPB

Sample number	FOM			
	Pure PTP	PTP+0.05mass.% DPB	PTP+0.1mass.% DPB	PTP+0.3mass.% DPB
1		1.22	1.17	1.15
2		1.05	0.88	0.92
3		1.44	1.09	1.18
4		1.45	1.20	1.13
5		1.13	1.32	1.16
6		1.33	1.21	—
	1.33			

Such results previously can be explained by following considerations. As it was mentioned earlier, the greater slow component contribution to total scintillation pulse, the higher the selective ability of scintillator (FOM-value). The slow component forms in process of triplet-triplet annihilation, which, in its turn, is regulated by the processes of transport and diffusion of triplet-excited states. Also, the molecules of dopant (DPB) are close in size to the host molecules (PTP), but are not identical to them. So, it can be supposed, that DPB molecules create inhomogeneities in PTP lattice which are able to influence on transport triplet-excited states and even disturb it at sufficiently high concentrations. It requires carrying out feather studies with higher concentrations of DPB, which was initially added into the melt, to verify this suggestion.

CONCLUSIONS

The FOM-values characterizing the pulse shape discrimination ability of a scintillator were obtained for PTP single crystals, containing different amount of DPB, under mixed alpha+gamma radiation. The content of DPB in PTP crystals varied from 0 (pure PTP) to approximately 0.8 mass.%.

Obtained results show, that with increase of introduced DPB concentration the pulse shape discrimination ability of PTP single crystals is getting worse in general, but do not allow to distinguish a clear dependency. Presumably, such results can be explained by suggestion that DPB molecules create inhomogeneities in PTP lattice which are able to influence on transport and efficiency of recombination of triplet-excited states. However, to proof such suggestion it is necessary to carry out additional investigations with samples, obtained with higher initial concentrations of dopant in the melt.

In the investigated concentration range, a tendency for weak growth in light transmittance is observed as the DPB content in PTP single crystals increases.

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ЗДАТНІСТЬ ДО РОЗДІЛЬНОЇ РЕЄСТРАЦІЇ ЗА ФОРМОЮ ІМПУЛЬСУ МОНОКРИСТАЛІВ *p*-ТЕРФЕНІЛУ, АКТИВОВАНИХ РІЗНОЮ КОНЦЕНТРАЦІЄЮ 1,4-ДИФЕНІЛ-1,3-БУТАДІЕНУ

А.В. Креч, І.В. Лазарєв, С.В. Махота, Є.В. Мартиненко, О.А. Тарасенко

Досліджувався вплив концентрації активатора на здатність до роздільної реєстрації за формою імпульсу монокристалів *p*-терфенілу, активованого 1,4-дифеніл-1,3-бутадієном. Визначено величини показника якості (FOM) для монокристалів *p*-терфенілу, які містили різні концентрації 1,4-дифеніл-1,3-бутадієну, при змішаному альфа+гамма-опроміненні. Використовувався метод зарядово-часової трансформації. Вміст 1,4-дифеніл-1,3-бутадієну в кристалах *p*-терфенілу змінювався від 0 (чистий *p*-терфеніл) до приблизно 0,8 мас.%. Отримані результати показали, що зі збільшенням концентрації введеного 1,4-дифеніл-1,3-бутадієну здатність до роздільної реєстрації за формою імпульсу монокристалів *p*-терфенілу загалом погіршується.